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PROCEDURES FOR THE SYSTEMATIC SEPARATION AND ANALYSIS OF RADIOACTIVE PARTICLES FROM NUCLEAR DETONATIONS

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Contents

PROCEDURES FOR THE SYSTEMATIC SEPARATION AND ANALYSIS OF RADIOACTIVE PARTICLES FROM NUCLEAR DETONATIONS

Abstract

A set of procedures for the systematic separation and analysis of particle groups has been developed for use in a reexamination of early time aerial filter and local fallout samples of radioactive particles from nuclear detonations.

The radioactive particles produced by nuclear detonations have radioisotopic compositions which vary with both particle size and type. To determine how individual radionuclides are distributed among the particles, it is first necessary to group the particles according to size and/or type. For the most part, conventional particle analysis techniques do not provide the required separated particle fractions.

The range of particle diameters encountered extends from less than $0.01\ \mu$ up to several cm. No single separation technique is effective over this entire range. However, all samples fall into one of three narrower range categories for which single procedures do apply. Thus, for all local fallout samples, the majority of particles are in the $100\text{-}\mu$ to few-cm diam range. Such samples are size separated by a modified dry sieving technique and density separated by flotation. All aerial filter samples from surface and near surface detonations have particles predominantly in the 1- to $100\text{-}\mu$ diam range. These samples are size separated by gravitational sedimentation in stabilized fluid columns which are open at the base so that discrete size fractions can be collected according to time of sedimentation. Density separation is accomplished by density gradient centrifugation. The third category of samples, which includes all aerial filter samples from airbursts, contains particles whose size range is principally between 0.01- and $1.0\text{-}\mu$ diam. These are subjected to size separation and density determination by centrifugal sedimentation in stabilized, open bottom, fluid columns.

The separated particle groups from all categories of samples are subjected to physical examination and gamma spectrometric analysis for the purpose of determining radioisotopic and mass distribution as a function of particle size for each class of radioactive particle produced by the detonation.

Introduction

The radioactive particles produced by nuclear detonations constitute the single most important source of man-made radiation in the biosphere. The nature and extent of interaction of the particles with a particular biological system depends ultimately on the surface density of deposition at the point of interaction, the size distribution of the particles deposited, and the physical, chemical, and radiological properties of the particles. Particle transport through the atmosphere from a point of origin to a point of deposition depends on particle settling rate and hence on particle size. Therefore, assessment of the overall biological effect of a particular nuclear detonation requires that for the complete particle population, we know the size distribution, the radioisotopic composition as a function of size, and finally, the chemical and physical properties of the particles, again as a function of size. The experimental procedures described here were developed in order to provide this information.

Discussion

Previous experimental work in this field has, with few exceptions, been of limited applicability to the total problem. Thus, although innumerable particle size analyses of fallout material have been made, they apply only to a particular sample and cannot, in general, be related to the whole population. Nor, do such analyses give the radioisotopic composition of the particle fractions studied. Similarly, a considerable amount of work has been reported in which the physical and chemical characteristics of individual fallout particles are determined; but here again the relationship of the particles analyzed to the whole population cannot be established. An extensive critical compilation of results obtained prior to 1962 has been prepared under sponsorship of the Defense Atomic Support Agency.[1]

Two experimental studies which do treat the entire population have been published. An Air Force Weapons Center document[2] reports size and radioisotopic distribution for a low-yield land-surface detonation; a British study[3] reports size distribution, chemical composition, and general radiation characteristics of the particle population from a low yield airburst.

A further source of relevant information lies in the diagnostic radiochemical data obtained by Lawrence Radiation Laboratory, Livermore, and Los Alamos Scientific Laboratory on close-in aerial filter and fallout samples. These data exist as a series of classified reports listed according to the individual nuclear test to which they pertain. Data obtained by the U. S. Naval Radiological Defense Laboratory has been used to determined how individual radionuclides fractionate under various detonation conditions. These results are reported by Freiling.[4]

The relative abundance of radionuclides found in a given particulate sample from

a nuclear detonation is in general different from the relative abundance of formation and from the relative abundance observed in other samples. This effect which is termed fractionation, is principally due to the fact that radionuclides of refractory elements condensed early and radionuclides of volatile elements condensed late with most elements exhibiting an intermediate behavior. In general, the overall size distribution of particles in the condensing system changes with time so that different radionuclides are distributed differently with particle size. Particle samples collected at different locations at various times are inherently biased with regard to the size of particles collected, so that different samples exhibit different radionuclide composition. A few radionuclides, those of elements which can exist in the vapor state for very long times such as long lived krypton-85, carbon-14 as carbon dioxide, or tritium as water vapor are only partially scavenged. All other radionuclides, with the possible exception of radioiodine, appear to be quantitatively scavenged by the particle population.

Based on information cited above, and on results obtained in the present program, it is possible to categorize nuclear detonations according to the particle populations they produce.

AIR BURST

A detonation which occurs more than one fireball radius above the surface is considered an airburst. The particles, produced by condensation from the vapor state of the metallic constituents of the device, form a single class of metal oxide spheres. Particle sizes range from about 0.01- to 20-μ diam, and particle density approximates that of the metal oxide. Radionuclide fractionation behavior results in a straight line relationship on a log-log plot if, for successively later air samples, the observed atom ratio of isotope A to refractory isotope R is plotted against the ratio of isotope B to refractory R. That is, the interisotopic atom ratios for such a sample series can be expressed:[5a]

$$A/R = (B/R)^{n_A} \qquad (1)$$

where A/R and B/R are atom ratios for isotopes A, B to refractory species R, and where n_A depends on detonation conditions.

LAND SURFACE BURST

A detonation occurring on a land surface, or less than one fireball radius above the surface and not supported by a metal tower, is considered a land surface burst. Such detonations produce particle populations which clearly consist of two dominant components - (1) crystalline particles and (2) "glass" particles produced not by condensation but by partial melting and resolidification of soil material. The crystalline particles which are also local soil material, enter the radioactive cloud at a late time and hence are not melted. Spherical metal oxide particles characteristic of airbursts constitute an insignificantly small proportion of the population. The size range of the overall population is from a few tenths of a micron to a few hundred microns diameter, with the "glass" particles

contributing principally to the larger diameter range and the crystalline to the smaller. Crystalline particle densities are those of the local soil material. Glass particle densities are equal to or slightly less than those of the local soil. Radionuclide fractionation behavior in surface bursts[5b] has been treated similarly to that in airbursts, however, there is evidence in the present work as well as in earlier work[6] that the data for all samples may be better fit by a series of linear relationships of the form

$$A/R = a + b(B/R) \qquad (2)$$

where again A/R and B/R are atom ratios and a and b are constants for set for a given isotope set, for a particular detonation.

TOWER BURST

A detonation conducted above a terrain surface on a metal tower is a tower burst. If the energy of the detonation is sufficient to vaporize the entire tower mass, the particle population produced is like that described under "Land Surface Burst." If, however, the entire tower is not vaporized, the overall particle population will consist of three components: the crystalline and "glass" components described under Land Surface Burst plus a metal sphere population which arises from melted (not vaporized) tower materials resolidifying as spheres. Such spheres are predominantly metallic rather than metal oxide, and exhibit the density and magnetic properties of the tower material. The size range of the

spherical component is from a few microns to perhaps a few hundred microns diameter. Radionuclide fractionation can be described by a series of linear relationships of the form:

$$A/R = a + b(B/R) + c(C/R) \qquad (3)$$

where A/R, B/R, and C/R represent atom ratios of isotopes A, B, and C to refractory species R; and a, b, and c are constants for a particular detonation.

LAND SUBSURFACE BURSTS

An underground detonation which produces a fireball above the surface is designated a Land Subsurface Burst. The particle population in the cloud resulting from such a detonation has the two components — crystal and "glass" — described under Land Surface Burst. A third component, which also has a glasslike character, may be observed in the very close-in fallout samples. This component appears to result from soil material which interacted with the fireball at high temperature but which was separated from the fireball early, before the temperature had fallen below the melting point of the soil materials. The particles in this component have diameters ranging from tens of microns to several centimeters and have densities which are likely to be quite low compared to those of the original soil components. The relative abundance of radionuclides in this component is fairly constant from sample to sample and thus appears to be independent of particle size. If we indicate by subscript 1, 2 the aerial cloud

population and by subscript 3 the third particle component described above, the radionuclide distributions can be best represented by two sets of equations:

$$A_{1,2}/R_{1,2} = a + b(B_{1,2}) (R_{1,2})$$

$$A_3/R_3 = c(B_3/R_3)$$

(4)

VENTED UNDERGROUND BURST

An underground detonation which produces no visible fireball but which results in the release of radionuclides through fissures or other vents is designated a vented underground burst. Such an event produces a single particle class — ambient air particles, crystalline in form with condensable radionuclides deposited on their surface. The particles range in size from about 0.01 μ to perhaps 10 μ and have a density which is characteristic of the ambient particles. Radionuclide abundance is independent of particle size.

WATER SURFACE BURST

A detonation which occurs at or near the surface of the open sea and which produces a visible fireball is designated a water surface burst. In those cases where the detonation platform is completely vaporized, the basic particle population produced is like that described under airburst. However, here the situation is complicated by the formation of so-called "slurry" particles. These consist of condensed sea salts which first scavenge spherical particles by agglomeration and then eventually decompose by hygroscopic action. The radioactivity however is primarily associated with the metal oxide spheres of the basic population.

In a case where the energy of the detonation is insufficient to vaporize the platform, the particle population will differ from that of an airburst, by containing spherical metallic particles (and even crystalline, glassy components, if soil-like ballast has been used).

DEEP SUBSURFACE DETONATION

The class of detonations which are completely contained includes both underwater and underground detonations. These introduce no radioactivity into the atmosphere and are not included in the experimental program described herein.

The experimental program has a twofold objective: (1) to determine the specific abundance (atoms/mg) of the individual radionuclides as a function of particle size and class, and (2) to determine the size distribution and relative abundance of each of the particle classes. This requires that particle samples encompassing all particle classes and sizes he subjected to size separation, class separation, weighing, and radioisotopic analysis. Since numerous long-lived radioactive species are produced by nuclear detonation, it is possible to analyze particle samples of considerable age.

The principle sources of samples for the present program are residual portions of aerial and fallout samples collected at the time of detonation (for radiochemical diagnostic analyses). The long range objective of the program is to determine how the particle distribution functions for each type of event change with the yield

and mass of the device detonated. The overall program has required development of procedures for separation and analysis of all classes and sizes of particles. No one method of separation is effective for the entire range of sizes encountered. However, it is possible to cover the entire range with three procedures. Thus, local fallout samples, aerial filter samples from land surface and near surface detonations, and aerial filter samples from airburst detonations all present different particle size ranges which must be treated differently. The procedures used for each of these cases are described in the following paragraphs.

Separation Procedures — Local Fallout Samples

The principle reason for examination of local fallout samples is to obtain size and radioisotopic composition data for those particle groups which were no longer present in the cloud at the time of aerial sampling. In the case of land subsurface detonations this part of the particle population may represent a very high percent of the total. In the case of land surface detonation, the part of the population missing from the cloud may or may not be significant depending on aerial sampling times and locations. In the case of airburst detonations the fraction missing from the cloud at sampling time is negligible.

All fallout samples of interest to this program were collected on open trays and transferred at the point of collection to sealed containers. The sample may range from a few grams to several hundred grams in size. For such samples, the size separation is most effectively accomplished by dry sieving through a series of graded screens. The technique of dry sieving and its limitations have been adequately described elsewhere.[7a] In the present work, a U.S. Standard sieve series with 710-, 500-, 350-, 250-, 177-, 125-, 88-, and 62-μ openings is used. These are stacked and subjected to mechanical shaking for a 6-hour period, the fraction less than 62 μ is caught in the cup at the base of the stack. Depending on sample size, commercially available wire-woven sieve sets of 3- or 8-in. diam are used. A Cenco-Meinzer Sieve Shaker, one of several commerically available models, is used to accomplish the sieving.

One undesirable feature of dry sieving as a particle separation technique is that fine particles tend to stick to the sieved large particles. In general, such "fines" are substantially less than the minimum sieved size of 62 μ and can be separated from the larger particles by washing in an appropriate non-reactive fluid. Carbon tetrachloride, CCl_4, which has no detectable solvent effect on any of the radioactive particle types encountered in the examination of past events, is used as the washing medium. However, in the samples examined, no radioiodine was present and in fresh material radioiodine might be leached. The particles collected in a given sieve are transferred to a 150-ml beaker and covered with CCl_4 to a depth of 1 in., and the suspension is swirled briefly and allowed to settle for whatever minimum time is required to allow the

sieve fraction particles to settle out. The supernate is then immediately decanted into a larger beaker and is later combined with the supernates from two additional, identical washings. The CCl_4 may be evaporated from the supernate suspension, and the particles in it combined with the less than 62-μ fraction. Microscopic examination of the sieve fraction particle can be used to monitor effectiveness of the washing, which is repeated for each of the sieve fractions.

Close-in, open tray fallout collections are likely to contain inactive material resulting from the transport of local soil particles that have been raised up at detonation as a result of shock and wind effects. An accurate assessment of size distribution and isotope specific activities for the radioactive particles requires that the contribution of these inactive particles be accounted for. This may be accomplished by a density separation. Especially for the larger particle sizes such as are found in local fallout, radioactive particles which have been subjected to melting and resolidification are likely to contain voids and thus to have densities significantly lower than that of the original soil material. Therefore, by shaking the mixed particle sample in a fluid whose density is less than that of the least dense soil component, it is possible to obtain a fraction which contains only radioactive particles. For each of the washed sieve fractions the procedure is the same. The entire sieve fraction is weighed and transferred to a vial and its gamma counting rate determined in a well crystal. Either the entire fraction or a 200-mg portion (whichever is smaller) is transferred to a 10-ml plastic centrifuge tube. Fluid of appropriate density is used to fill the tube to within 1.5 cm of the top. For silicate soils, a solution of density 2.5 g/cm^3 is made by mixing 30-vol % CCl_4 and 70-vol % bromoform, $CHBr_3$.

Bromoform exhibits no solvent action on any of the radioactive particle types encountered in old samples. (Again, radioiodine may be dissolved to some extent from fresh particulate material. In this case, gross gamma counting would have to be replaced by gamma spectral analysis for individual radioisotopes.) The tube is stoppered and shaken vigorously for a few seconds and then set aside for several hours to allow the higher density particles to settle out. Using a tube slicer such as that available from the Beckman Instruments Co., the tube is cut about 1 cm from the base and the two fractions are separately filtered onto 1-μ pore size, 1-in. diam, weighed membrane filters, and are washed with CCl_4, dried, and weighed. The filters are then gamma counted in the same well crystal geometry as the original sieve fraction was counted. The mass of radioactive particles in the original sieve fraction may be calculated:

Mass in original fraction

$$= \frac{\text{Gamma counts/min in original fraction}}{\left(\dfrac{\text{Gamma counts/min}}{\text{Mass}}\right)_{\text{light particles}}} \quad (5)$$

This assumes that the specific activity observed for the light particles is representative of all radioactive particles in the sieve fraction. Typical results which indicate that this may be a valid assumption, show that perhaps 90-95% of the activity goes into the low density fraction. A ratio of 10 to 1 in specific activity

between the low and high density fraction is not unusual.

The final step in the separation procedure is to mount the filters containing the particle fractions for gamma spectrometry. Each sample is mounted in a 1-in. i. d. , polyethylene cup, 1/2 in. high, and capped with a 1-in. lucite disk as indicated in Fig. 1. The lucite disk is sealed to the polyethylene wall with Duco Cement or its equivalent. The mounting method used is
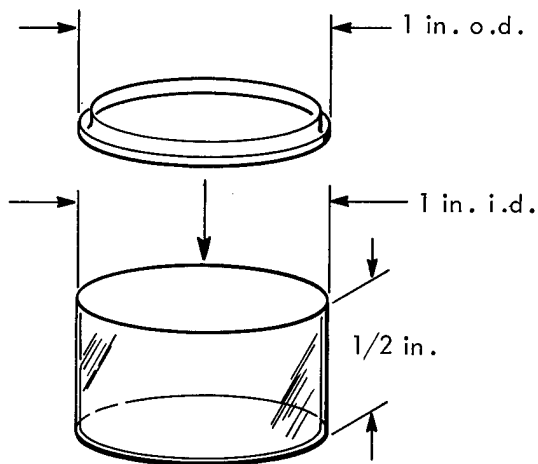

Fig. 1. Sample mounting cup for gamma spectrometry.

intended to provide the maximum reproducibility of geometry, together with a minimum likelihood of detector contamination. Gamma spectrometric counting procedures are the same for all types of samples and are described briefly following discussion of the other separation procedures.

SUMMARY—FALLOUT SAMPLES

(1) Samples are dry sieved, with successive sieve sizes varying by a factor of $\sqrt{2}$.

(2) Each sieve fraction is washed with CCl_4 to remove fines. These are recombined with the fine fraction.

(3) Each washed sieve fraction is weighed, counted, and subjected to a density separation in order to isolate radioactive particles from the gross sample.

(4) Density separated size fractions are weighed, counted, and mounted for gamma spectroscopy.

Separation Procedures — Aerial Samples from Surface Bursts

Surface bursts in the context of this discussion are intended to include all cloud-producing categories of nuclear detonations except airbursts. The particle size range of surface bursts is principally in the 1- to 100-μ diam region while that of airbursts is in the 0.01- to 1.0-μ range.

All aerial samples of interest to the program were collected on IPC type 1478 filters. These are made from cellulose fibers impregnated with a plasticizer (Kronisol, the dibutyl ester of pthallic

acid) and backed with a scrim made of cellulose fiber impregnated with calcium sulfate. The IPC 1478 filter is used because it has a high retention efficiency at high flow rates. The high retention efficiency has, for particle analysis purposes, the undesirable effect that particles once entrapped cannot be separated from the filter medium. In order to recover the particles it is necessary to destroy the filter. However, this destruction must not produce any alteration in the

physical, chemical or radioactive character of the particles.

The method of low-temperature ashing which employs oxygen gas at low pressure and low flow rate excited in an rf field to produce reactive oxygen was used with some success in the earliest phases of the program.[8] However, it proved ineffective for filter material which was manufactured prior to 1962 and which had a significantly higher inorganic contamination. Further, coral island source samples showed variable percentage losses of volatile radioactive species in the ashing process and airburst samples showed large variable losses of all isotopes. For these reasons, the use of this technique was discontinued. In its place, ashing in a muffle furnace at 400°C for 4 to 7 hr is used to destroy the filter material. The ashing is done in a Pyrex petri dish; a higher-walled vessel hinders the ashing process. The conditions are quite critical—with lower temperature or shorter time there is a filter residue; at higher temperatures and longer times, there is a loss of volatile radioactive species. At 400°C, the only long-lived radioisotope lost at all is cesium-137, which exhibits a maximum loss rate of 0.1% per hour of ashing. Ashing is completed when the black discoloration, which appears after a few minutes ashing, is completely gone. The time required for complete ashing depends somewhat on filter paper size. A 1-in.^{2} filter ashes in 4 hr; a 6-in.^{2} filter, the largest used in the program, requires about 7 hr, although occasionally a longer period is needed. The weight of the original filter is approximately 100 mg/in.^{2}; the weight of the residue ranges from 0.04 to 0.08

mg/in.^{2}. Observed surface densities of particles on exposed papers for surface detonations range from 2 to 10 mg/in.^{2}; for subsurface bursts, values as high as 120 mg/in.^{2} have been found. Thus for the general category of surface detonations, the filter residue constitutes a negligible fraction of the particle sample weight. (In all cases, the scrim backing is removed before ashing.)

SIZE SEPARATION PROCEDURE

For particles less than 60 μ in diameter dry sieving does not produce effective size separation because of particle agglomeration. The sieving range can be extended down to perhaps 20 μ by wet sieving, but at smaller size, agglomeration is still a problem. Size separation down to the 1-μ region can best be accomplished by gravitational sedimentation in a fluid medium. The rate at which a particle settles in a viscous medium is given by Stokes' Law, one form of which may be expressed:[7b]

$$\frac{dz}{dt} = g\alpha M^2 \frac{(d - \rho)}{18\eta}$$ (6)

where:

  z = Vertical distance
  t = Time
  g = Gravitational constant
  α = Particle shape factor
  M = Size of particle, averaged over all possible orientations of settling
  d = Particle density
  ρ = Fluid density
  η = Fluid viscosity

When z is expressed in cm; t in min; M in μ; d and ρ in g/cm^3; and η in cP, the above expression reduces to

$$\frac{dz}{dt} = 3.27 \times 10^{-3} \, \alpha M^2 \frac{(d - \rho)}{\eta} \qquad (7)$$

For spherical particles, the particle shape factor α is identically equal to 1, and M is the diameter. For particles whose maximum-to-minimum diameter ratio is less than 4, α can be taken as equal to 1, since particles of this type do fall at very much the same velocity as spheres of the same density and volume.[9] In all that follows, α will be taken equal to 1, and M will be referred to as "particle diameter."

The fact that the time required for a particle to fall a given distance is inversely proportional to (particle diam)^2, makes sedimentation in a viscous fluid an extremely effective method for particle separation. However, to obtain useful results it is essential that convective motion of the column fluid be eliminated. This is best done by establishing a suitable density gradient in the column. Such a gradient is established by mixing two solutions in continually varying proportions so that the base of the column is at a density of 0.914 g/cm^3 and the top at 0.907, with the density intermediate heights changing smoothly between these extremes. A mixture of 6-vol % CCl$_4$ and 94-vol % Dow-Corning silicone fluid (2-cSt viscosity grade) is used as the high density component; and a 5-vol % 95-vol % mixture of the same two fluids is used as the low density component. Neither fluid has any detectable solvent effect. The density gradient imposed corresponds to perhaps 10° temperature difference from top to bottom and is sufficient to completely suppress vertical convective motion. The apparatus used for preparing the continually varying mixture is described by Oster and Yamamoto.[10] It is shown in Fig. 2.

To use gravitational sedimentation for the physical separation of particle groups, it is necessary to start the sedimentation from a narrow layer at the top of the column and to periodically remove accumulated particles at the base of the column. The apparatus developed for this purpose is illustrated in Fig. 2. The base of the column is 24-mesh, 7.5-mil stainless steel wire screen held in place by a stainless steel retainer ring, joined to the glass column by an epoxy resin seal. The glass column is 6-cm i.d. at its base and 31 cm in length from the base to where it narrows down to a 4.2-cm-i.d. neck which is 4 cm in length and terminates in a 40/50 ground glass joint. In earlier work a straight cylindrical column 6-cm i.d. and of equivalent length was used. However, in the present configuration, the particles, which are introduced in a fluid layer on top of the 31-cm column, are farther from the cylinder wall for most of the sedimentation period; therefore, losses to the wall are minimized.

The procedure for using the column is as follows:

(1) Filling. The column, clamped to a metal rack, is sealed at the base with a large rubber stopper, covered with a sheet of 1-mil Mylar and pressed in place by a lab-jack. The column is filled, using 500 ml of 6-vol % carbon tetrachloride in the left tube of the density gradient apparatus and 509 ml of 5-vol % in the right. The fluid is introduced at the base of the column by means of a polyethylene tube. Since the first drop over is the
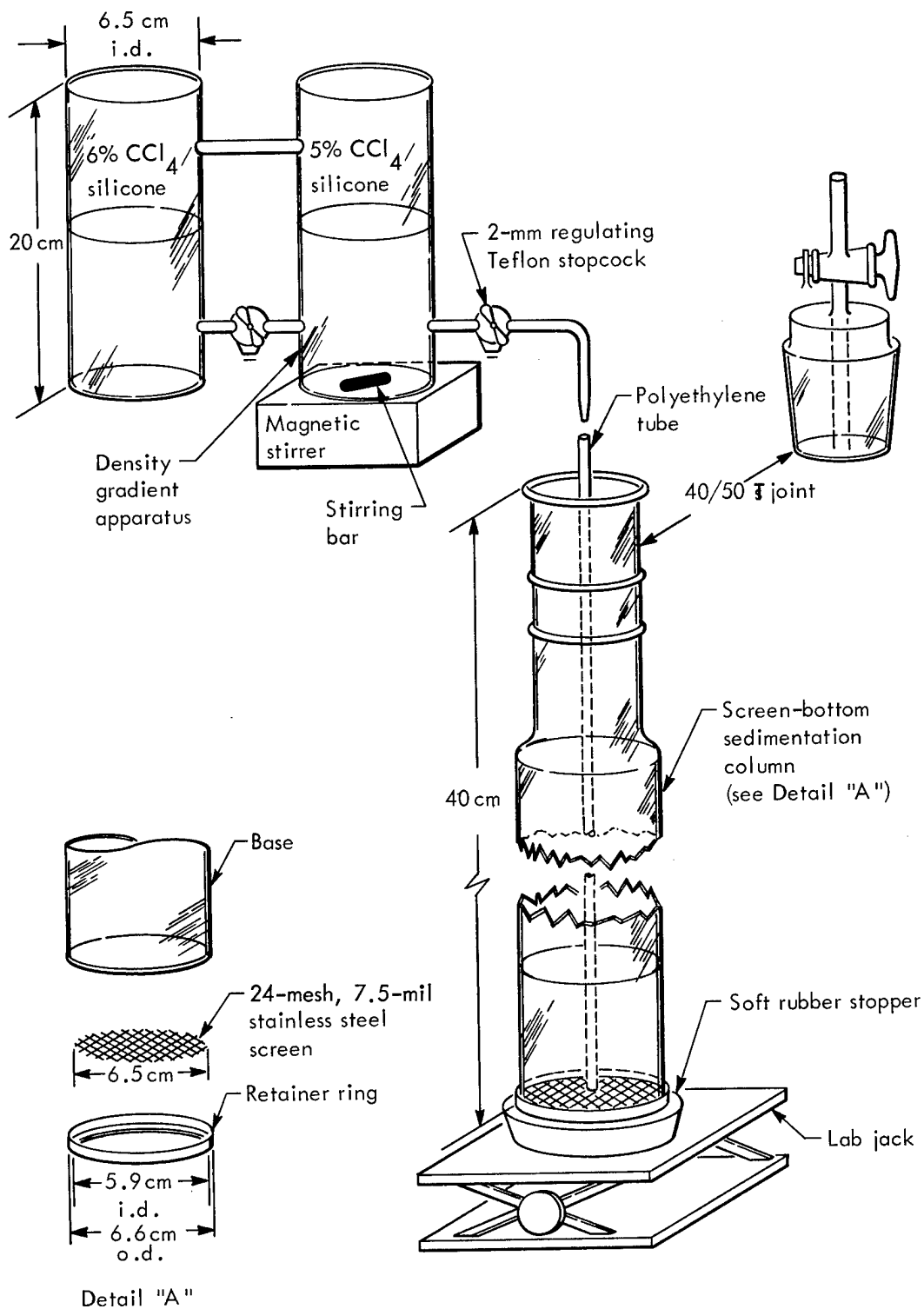
6.5 cm i.d.

6% CCl$_4$/silicone

5% CCl$_4$/silicone

20 cm

2-mm regulating Teflon stopcock

Density gradient apparatus

Magnetic stirrer

Stirring bar

Polyethylene tube

40/50 $\mathfrak{F}$ joint

Screen-bottom sedimentation column (see Detail "A")

40 cm

Base

24-mesh, 7.5-mil stainless steel screen

6.5 cm

Retainer ring

5.9 cm i.d.

6.6 cm o.d.

Soft rubber stopper

Lab jack

Detail "A"

Fig. 2. Apparatus for preparing of a density gradient for gravitational sedimentation.

lightest, the fluid present floats on that which is entering. The column is filled to a height of 31 cm and the filler tube is removed.

(2) _Dispersion of Particles._ In order to effect particle separation in a fluid medium it is essential that the particles be individually suspended in the fluid. Particle samples as received are agglomerated and contain entrapped air which tends to maintain the agglomeration. Dispersion of the particle sample into the fluid is accomplished by first boiling the suspension to drive off entrapped air and then subjecting the suspension to prolonged rolling in a sealed vial.

To the particle sample in a 150-ml beaker is added 20 ml of 2-cSt silicone

sleeve whose i. d. is slightly less than the common o. d. of the vial and stirring motor chuck (see Fig. 3). The stirring motor is operated at low speed in a horizontal position. The resultant rolling of the fluid suspension over the vial wall slowly disperses the particles. Rolling of the fluid suspension is carried out for a minimum of 16 hr and is continued until the sample is added to the top of the sedimentation column.

(3) _Addition of Sample._ The dispersed particle sample is poured from the vial, down a stirring rod, and onto the top of a previously prepared column. An additional 20 ml of 2-cSt silicone fluid are used to wash the vial walls and insure a complete
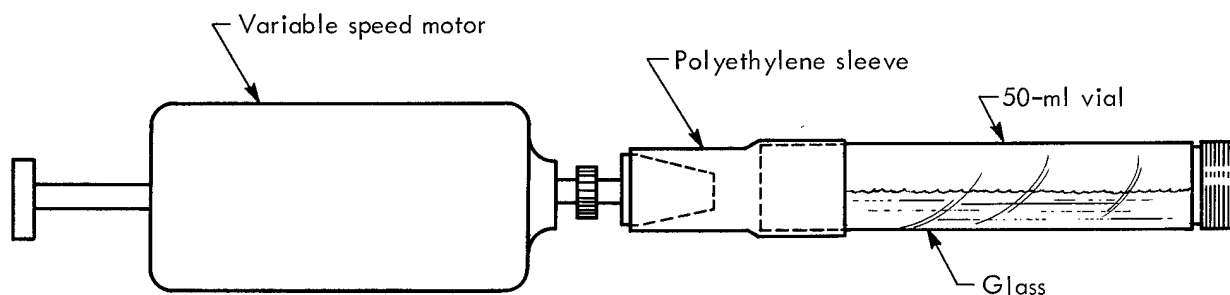

Fig. 3. Sample blender.

fluid containing 0.1% wetting agent (Aerosol OT). The beaker is placed on a hot plate and heated to boiling (210°C). The suspension is boiled vigorously for 30 min and then allowed to cool. The cool suspension is transferred to a 50-ml glass screwcap vial and 10 ml of 2-cSt silicone fluid is used to wash the beaker. The screwcap is fastened and secured in position by means of plastic tape, and vial is attached to the chuck of a variable speed stirring motor by means of a polyethylene

particle transfer. The column dimensions and reagent volumes specified will permit particle samples of up to 200 mg to be dispersed and separated. Average particle density is in the vicinity of 2.5 g/cm^3 and the silicone fluid density is 0.871 g/cm^3. Therefore the density of the suspension which is poured onto the column is less than 0.877 g/cm^3. Since this density is significantly less than that at the column top, the particle suspension will float as it is added.

(4) Sealing the column. With stopcock open and with the ground glass wall lightly greased, the inner part of the 40/50 joint is placed in position and the stopcock is closed. By lowering of the lab-jack, the mylar covered rubber stopper at the base of the column is removed. In its place is put a 300-ml beaker containing 10 ml of a 10-vol % CCl_4, 90-vol % 2-cSt silicone fluid mixture. The beaker is raised into position so that the column fluid and beaker fluid join. During the changeover period, the column is supported by surface tension at the screen. When the beaker is in place, the column is supported by atmospheric pressure. A very slight outward bow of the screen is desirable in order that no air bubbles are trapped between beaker fluid and column fluid at the time of joining. In order for there to be no density streaming at the fluid interface the beaker fluid must be more dense than the column fluid. Particles reaching the base of the column fall into the collecting beaker. The collecting beaker can be replaced by another containing fresh 10-vol % CCl_4/ 90-vol % silicone fluid at any time so that physically separate particle size fractions can be obtained. Particles which may have fallen on the screen tend to be washed off by fluid agitation set up at the interface when fluid surfaces join or separate. The operations of sample addition and collection are shown schematically in Fig. 4.

(5) Particle fraction collection schedule. The relationship between particle size and time of arrival at the screen is given by:

$$tM^2 = \frac{h\eta}{3.27 \times 10^{-3} (d - \rho)} \qquad (8)$$

where h = total cm fallen in t min, and other symbols as previously defined. Although fluid density, ρ, and viscosity, η, are both changing with column height, the change is so small (less than 1% between extremes) that a single mean value for each of these for the column can be used. Values corresponding to 5.5-vol % CCl_4, which is the mean composition of the column, are:

$$\rho = 0.909 \text{ g/cm}^3$$

$$\eta = 1.686 \text{ cP}$$

Column height is 31.0 cm and the thickness of the sample layer is 3.6 cm. The particle fall distance from the midpoint of the sample layer to the screen is therefore 32.8 cm. Hence

$$tM^2 = \frac{16910}{(d - 0.909)} \qquad (9)$$

For any particular particle density, the foregoing gives the relationship between t and M. For a generalized treatment, a reasonable value for a surface detonation particle density may be taken as 2.55 g/cm^3 with a standard deviation of 0.15. If d = 2.55 ± 0.15 g/cm^3 and ρ = 0.909 g/cm^3 then

$$tM^2 = \frac{16910}{1.64 \pm 0.15} = 10310 \pm 940. \qquad (10)$$

Hence M = 106 ± 5/t$^{1/2}$ gives the diameter in μ of particles reaching the screen at t min. The time is measured from addition of the sample to the top of the column. The independent variable in particle size distribution function is usually not the particle diameter but the logarithm of the diameter. Therefore, consecutive samples taken for the purpose of determining the
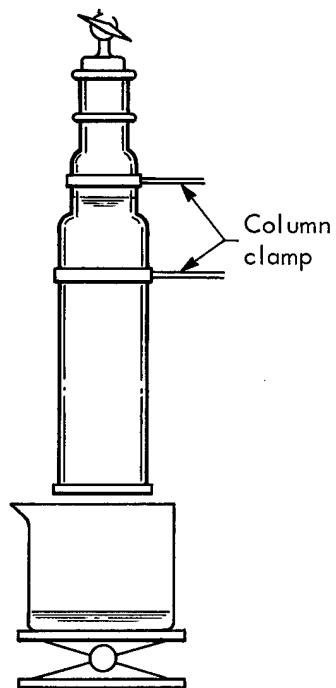
50-ml vial

Stirrer

Column clamp

32 cm

a. Sample addition

Column clamp
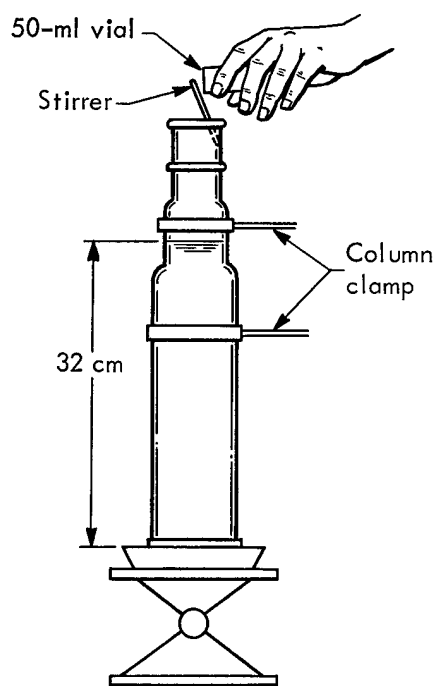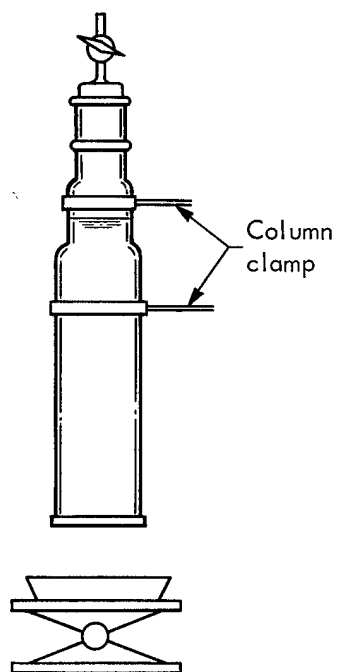
b. Column sealed, base support lowered

Column clamp
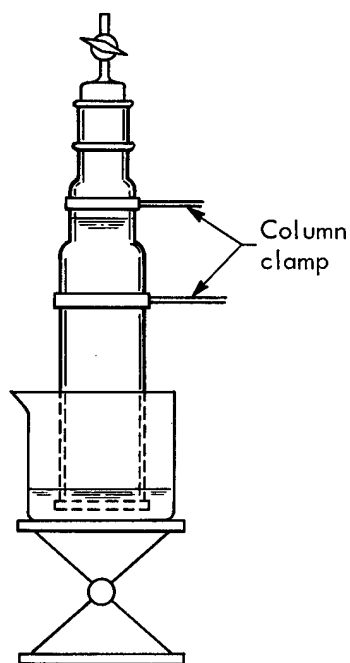
c. Particle collection beaker in place

Column clamp

d. Particle collection beaker removed

Fig. 4. The operations of sample addition and collection.

distribution should correspond to equal increments of the logarithm. For this reason sample collection intervals are based on a geometric series of time rather than an arithmetic one. Beakers are replaced at 2, 4, 8, and 16 min, and so forth out to 4096 min. Thus the first beaker contains particles greater than 73μ the second, particles between 73 and 51 μ, and so forth out to the one taken off at 4096 min which contains particles between 2.3 and 1.6 μ. At 4096 min, three additional fractions are obtained by draining the column. The bottom half, the next quarter, and the top quarter correspond to particles whose diameters are 1.6 to 1.1, 1.1 to 0.8, and less than 0.8 μ respectively. Draining is accomplished by slightly opening the stopcock at the top of the column; drainage is smoother if the collection beaker has fluid in it and is raised to a point where beaker fluid and column fluid are joined. The sampling procedure described produces a series of 15 particle size groups. The procedure is used routinely on all aerial filter samples from surface detonation.

(6) Column wash. In order to obtain quantitative recovery of the entire particle sample, it is necessary to police and wash the column wall. Particles lost to the wall usually constitute just a few percent of the total sample. However, the fraction lost from any given size group increases with decreasing size so that some bias is introduced into the measured distribution as a result of this effect.

(7) Processing of separated size fractions. Separated particle fractions are filtered onto weighed membrane filters (0.22-μ pore diam), washed with carbon tetrachloride, dried at 100°C, and weighed. The membrane filter is then mounted for gamma spectroscopy in a 1-in. polyethylene cup as described under fallout sample procedures. In cases where the particle fraction weight is small (i. e. , of the order of 1 mg) the membrane filter weighing method may prove unreliable. More accurate weight may be obtained after gamma spectroscopy is complete by ashing the sample in a pre-ignited, weighed porcelain crucible at 700°C, cooling, and weighing. At this temperature volatile radionuclides are lost but the inorganic oxide particle mass is unaffected.

DENSITY SEPARATION PROCEDURE

The radioactive particles produced by surface and near surface detonations exhibit physical and chemical characteristics which relate to those of the soil in the area of detonation. If a metal tower was present and not completely vaporized, spherical metallic particles from melted tower material will also be found. In general, soil derived particle densities will be in the range 2.2 to 3.2 g/cm^3, with the preponderance of particles having densities between 2.5 and 2.8 g/cm^3. Spherical metal particles have densities like that of the tower metal from which they were derived. For the density range of interest, it is possible to effect a density separation by density gradient centrifugation. In this technique, the particle sample is suspended on a fluid column having a continuously variable density covering a range of, say, 2.0 to 3.0 g/cm^3, and then centrifuged for a period

long enough to permit all particles to reach their equilibrium density level. The column may then be divided into density range fractions so that the suspended particles are separated according to density group. The stepwise procedure for density gradient separation is as follows:

(1) Preparation of the density gradient column. The density gradient column is prepared in a 50-ml round-bottom polyethylene centrifuge tube. The density gradient apparatus is a smaller version of the one previously described, and is shown in Fig. 5. With stopcocks A, B
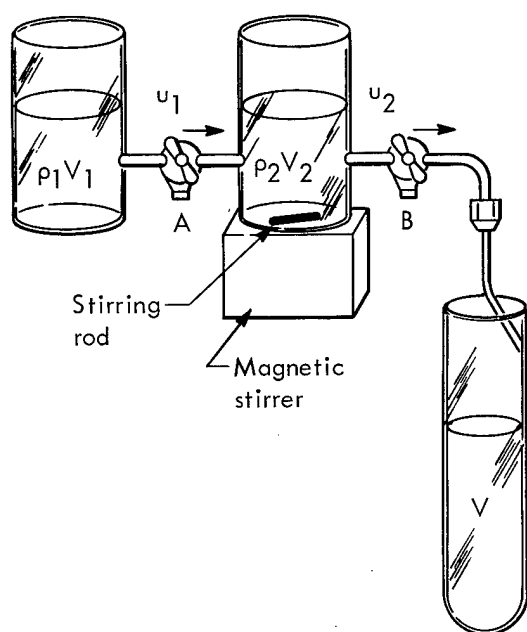
Fig. 5. Density gradient column for separation of particles from surface and near-surface detonations.

closed, heavy fluid is added to left side and light to the right. The initial densities and volume must obey the relationship $\rho_1^0 V_1^0 = \rho_2^0 V_2^0$ so that when the stopcocks are opened, the system will be in equilibrium. When the stirrer is turned on and stopcocks are opened, the flow is

as shown in the figure. The density of the fluid in the mixing chamber at any time, t, is given by[10]:

$$\frac{\rho_1 - \rho_2^0}{\rho_1^0 - \rho_2^0} = \left[\frac{(u_1 - u_2) t + V_2^0}{V_2^0} \right]^{(u_1/u_2 - u_1)} \quad (11)$$

where superscript zero indicates initial condition. However

$$u_2 t = V, \quad (12)$$

so that

$$\rho_2 = \rho_1^0 + (\rho_2^0 - \rho_1^0)$$
$$\times \left[\frac{V_2^0 - (1 - u_1/u_2)V}{V_2^0} \right]^{\left(\frac{u_1/u_2}{1 - u_1 u_2} \right)} . \quad (13)$$

But

$$u_1/u_2 = \frac{V_1^0}{V_1^0 + V_2^0} \quad (14)$$

so that

$$\frac{V_2^0 - (1 - u_1/u_2)V}{V_2^0} = V_2^0 - \left(\frac{V_2^0}{V_1^0 + V_2^0} \right) V$$
$$= 1 - \frac{V}{V_1^0 + V_2^0} . \quad (15)$$

And

$$\frac{u_1/u_2}{1 - u_1/u_2} = \left(\frac{V_1^0}{V_1^0 + V_2^0} \middle/ \frac{V_0^2}{V_1^0 + V_2^0} \right) = \frac{V_1^0}{V_2^0} ,$$
$$(16)$$

so that

$$\rho_2 = \rho_1^0 + \left(\rho_2^0 - \rho_1^0\right)$$

$$\times \left(1 + \frac{V}{V_1^0 + V_2^0}\right)^{V_1^0/V_2^0} . \quad (17)$$

However,

$$V_1^0/V_2^0 = \rho_2^0/\rho_1^0, \text{ and } V_1^0 + V_2^0$$

$$= V_{total} . \quad (18)$$

So we may write

$$\rho_2 = \rho_1^0 + \left(\rho_2^0 - \rho_1^0\right) \left(1 - \frac{V}{V_{total}}\right)^{\rho_2^0/\rho_1^0} \quad (19)$$

which gives the density of the mixture entering the base of the centrifuge tube as a function of the volume which has been transferred. This foregoing may be converted to a density-height relationship based on the centrifuge tube dimensions. For the present work, we have standardized two density gradients. Gradient I which covers the broad range 2.06 to 2.85 g/cm^3 is used to effect initial separations of components with widely varying

densities. Usually, a broad band of particles in the 2.5 to 2.7 g/cm^3 range is observed. Further separation of this band or separation of a sample of original material may be accomplished by use of the second gradient (gradient II) which covers the range 2.44 to 2.73 g/cm^3.

Both gradients are prepared from bromoform, 2-cSt silicone fluid mixtures. Composition and physical data for the heavy and light components are given in Table I. The density vs height relationships for the columns are given in Figs. 6 and 7.

The tube is filled to a height of 6.5 cm, which corresponds to a volume of 30.8 ml. The remainder of the 50-ml tube capacity is available for addition of the dispersed particle sample.

(2) Addition of Sample. A particle sample in a 30-ml beaker is suspended in 10 ml of 2-cSt silicone fluid containing 0.1% Aerosol OT. It is dispersed by boiling and rolling as described above. It is poured down a stirring rod onto the top of the density gradient column and the beaker is washed down with about 8 ml of 2-cSt silicone fluid. To avoid agglomeration, particle volume is kept below

Table I. Bromoform/silicone density gradient mixtures.

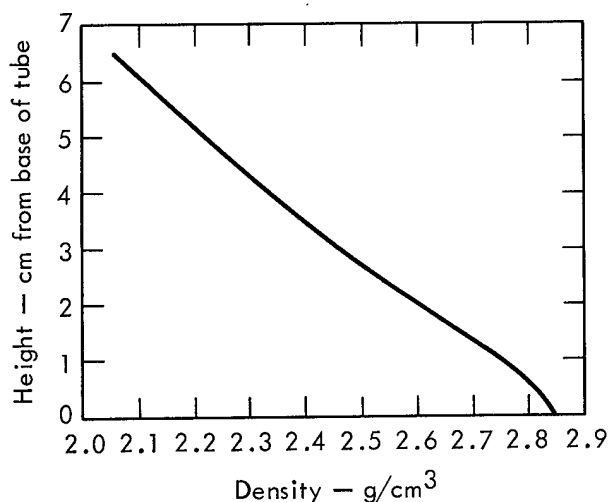|	Density gradient I		Density gradient II	
	High-density mixture	Low-density mixture	High-density mixture	Low-density mixture
Bromoform	100%	48%	93%	76%
2-cSt silicone fluid	0%	52%	7%	24%
Density (g/cm^3)	2.848	1.828	2.734	2.416
Viscosity (cSt)	1.93	2.01	1.90	1.96
volume (ml in density gradient apparatus)	19.5	30.5	16.0	18.0

Fig. 6. Density versus height for gradient I. (19.5 ml bromoform plus 30.5 ml 0.48 bromoform/0.52 2-cSt silicone in a 50-ml centrifuge tube filled to 6.5 cm from bottom.)
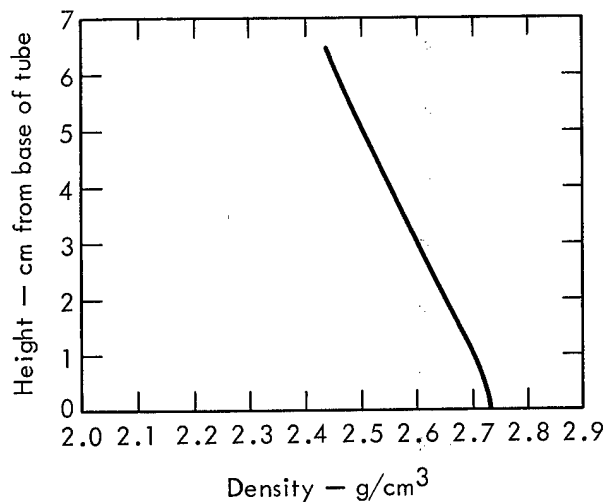

Fig. 7. Density versus height for gradient II. (16 ml 0.93 bromoform/ 0.07 2-cSt silicone plus 18 ml 0.76 bromoform/0.24 2-cSt silicone in a 50-ml centrifuge tube filled to 6.5 cm from bottom.)

0.5% of fluid volume. Thus particle sample size must be less than 125 mg.

(3) Centrifugation. Centrifugation is carried out at constant temperature in a refrigerated centrifuge with a swinging bucket head. Since centrifugation times are fairly long, it is necessary to maintain constant temperature during centrifugation in order to maintain constant density height relationship for the fluid. In the present work, the Sorvall Model RC-2B is used for centrifugation runs for density separations of soil type particles. Centrifugation time is set by the requirement that the smallest particle must reach its equilibrium density level. The rate of particle movement in a centrifugal field is given by

$$\frac{dr}{dt} = \frac{\alpha M^2 (d - \rho)\omega^2 r}{18\eta}, \qquad (20)$$

where:

r = Radial distance from axis of centrifuge

ω = Angular velocity,

and where other symbols are as previously defined.

When α is set equal to 1, and M is expressed in μ, r in cm, t in min, d and ρ in g/cm^2, and η in cP, Eq. (20) reduces to:

$$\frac{dr}{dt} = 3.655 \, M^2 \frac{(d - \rho)}{\eta} \, r$$

$$\times \left(\frac{rpm}{10,000}\right)^2 \qquad (21)$$

where rpm is the centrifuge speed in revolutions per minute. The relationship between centrifugation time, t, particle size, M, and particle density, d, is given by:

$$3.655 M^2 \left(\frac{rpm}{10,000}\right)^2 t = \int_{r_0}^{r_f} \frac{\eta(r)}{[d - \rho(r)]} \frac{dr}{r}$$

$$(22)$$

For gradient I in the sample region $5.3 < r < 7.3$, $\eta = 1.864$, and $\rho = 0.871$;

and in the density gradient column $7.3 < r < 14.3$, $2.00 < \eta < 1.93$, and $\rho(r)$ is as given in Fig. 6. The time required for a particle of density, d, to travel from the top of the sample layer to an equilibrium density level can be determined by numerical integration of the above expression. For equilibrium density level we take, $r_\alpha = r_\alpha - 0.1$; that is, it is taken as being 1 mm above that point at which the solution density equals particle density. The calculated values for gradient I are as follows:

Particle density g/cm³	$M^2 \left(\dfrac{rpm}{10,000}\right)^2 t$
2.2	1.61
2.4	1.74
2.6	1.71
2.8	1.64

Thus for centrifugation at 10,000 rpm for a particle of 1-μ diam, a maximum centrifugation time of 1.74 min is required. For smaller particles and lower centrifugation rates longer times are needed. In samples from surface detonations, the number of particles whose diameter is less than 0.1 μ is negligible. Therefore, at the 10,000 rpm rate, a time of

$$\frac{1.74}{(0.1)^2 (1)^2} = 174 \text{ min}$$

is sufficient to bring all particles to equilibrium.

Typical result of such a centrifugation is shown schematically in Fig. 8. Particle density groups can be separated by cutting (Beckman tube slicer) at the indicated levels and recovering the suspension. The intense band between cut 2 and cut 3 can be diluted with 5 ml of 2-cSt silicone
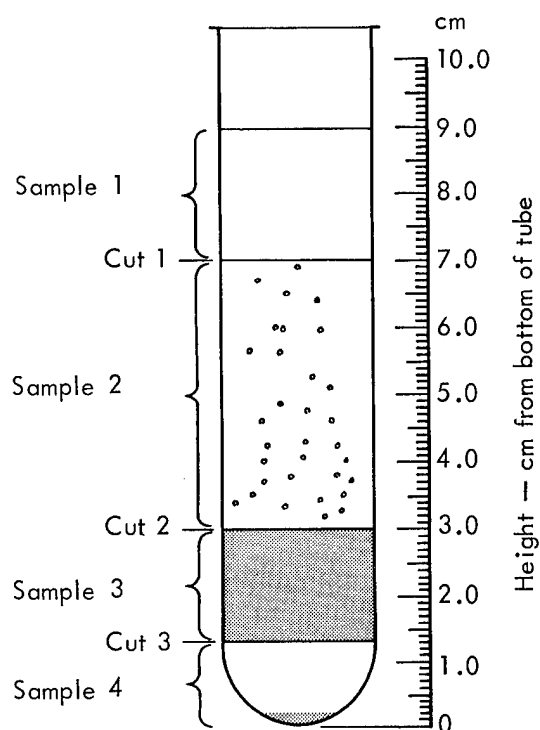

Fig. 8. Typical result of density separation of particles from ground surface burst. (Density gradient I.)

and transferred directly to the top of a density gradient column of type II for further separation. For gradient II,

$$\left[M^2 \left(\frac{rpm}{10,000}\right)^2 t \right]_{max} = 4.36$$

so that a total of 436 min centrifugation at 10,000 rpm is required to permit 0.1-μ particles to reach equilibrium density. Results typical of a feldspar/silicate soil separation are shown in Fig. 9.

(4) Processing of Separated Density Fractions. The separated density fractions are filtered, washed, dried, weighed, and mounted for gamma spectroscopy, microscopic examination or x-ray diffraction analysis as required.
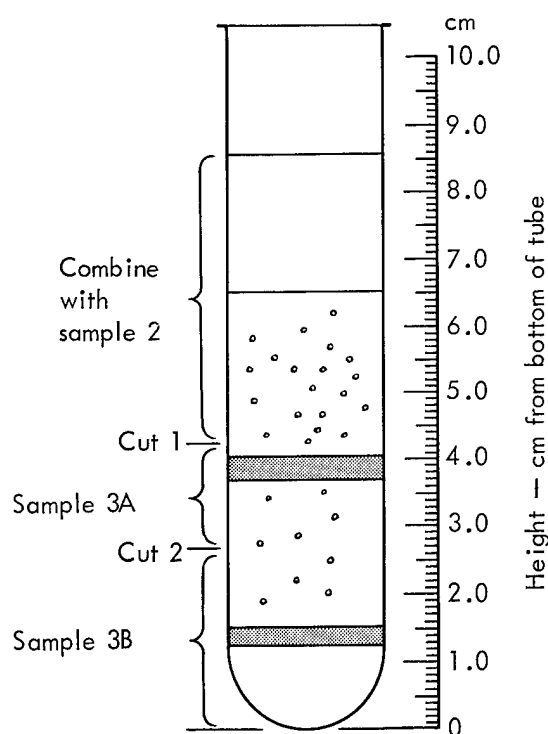
Fig. 9. Typical result of density separation of particles from 2.45- to 2.70-g/cm^3 density band. (Density gradient II.)

SUMMARY—AERIAL SAMPLES FROM SURFACE BURSTS

(1) Filter paper is eliminated by ashing 5 to 7 hr in a muffle furnace at 400°C.

(2) Particle sample is dispersed in 2-cSt silicone fluid containing 0.1% Aerosol OT by boiling followed by prolonged rolling.

(3) Size separation of samples up to 200 mg total is accomplished by sedimentation in a fluid column 32 cm in height stabilized against convective mixing by imposing a slight density gradient. Particle fractions are collected in beakers at the screen base of the column. Fraction collection beakers are replaced at 2, 4, 8 ... 4096 min, and the column drained in sections to give a total of 15 fractions corresponding to equal increments of the logarithm of particle diameter.

(4) Density separation of samples up to 125 mg total is accomplished by density gradient centrifugation. Density gradients covering the range 2.06 to 2.85 g/cm^3 and 2.44 to 2.73 g/cm^3 are used. Fractions are recovered by cutting the tube in sections.

Separation Procedures — Aerial Samples from Air Bursts

Particles which are produced by an air detonation constitute a unique separation and analysis problem. First of all, there is only one class of particle metal oxide spheres produced by condensation from the vapor state of the materials of the weapon and its ballistic case; second, the size range is lower by an order of magnitude than that observed for surface detonations; third, the weight of active particulate material collected in individual samples is smaller by perhaps 3 orders of magnitude than comparable samples resulting from surface detonations; and finally, the collections of particles obtained in the early air samples is in general completely representative of the entire particle population— in contrast to surface detonation samples, which are already depleted in large particles by the time of first collection by sampling aircraft. The fact that there is but one particle class and that a representative sample of it is available make the problem of

characterizing the radioactive particle population from an airburst conceptually simpler than that of surface bursts. However, low sample weight and small particle sizes make the experimental problem move difficult.

As in the case of surface detonations, all useful samples were collected on IPC Type 1478 filter paper. The filter paper ashing procedure described — 4 to 7 hr at 400°C in a petri dish in a muffle furnace — applies also to airburst filters. However, the 0.040- to 0.080-mg/in.^{2} residue from the filter is large relative to the particle mass, which is usually less than 0.010 mg/in.^{2}. During the ashing the filter shrinks continually until ashing is complete. The residue is a cluster of material somewhat spherical in shape with a diameter of 1/16 to 1/4 in. depending on the amount of filter ashed. The radioactivity is completely contained by this filter fiber skeleton. The petri dish in which the ashing was conducted is free of radioactivity when the fiber residue is removed. Gamma spectroscopy of the residue shows that activity recovery is quantitative for all isotopes. The fiber residue can be broken up by suspending it in a fluid and subjecting it to vigorous ultrasonic agitation. It is found that 20-kc ultrasonic agitation will accomplish the desired objective of separating the particles from the fiber residue without affecting the particles. A 20-kc agitation of soil type particles results in the breaking up of individual particles so that this ultrasonic frequency cannot be used for dispersal of surface detonation particles. However, the small metal oxide spheres of which airburst particle populations are made up are apparently unaltered by the

20-kc vibration. The procedure for separating the spherical particles from the fiber residue and dispersing them is as follows:

(1) The filter residue containing the particles is transferred to a 15-ml beaker and covered with 2 to 3 ml of either 2-cSt silicone fluid or carbon tetrachloride depending on what is to be done with the sample.

(2) The beaker is placed in the transducer coupled cup attachment of a commercial 20-kc ultrasonic generator and immersed in distilled water to the level of the interior liquid. Ultrasonic agitation is carried out for 30 min at an indicated power level of about 60 W. That is, the generator, which has a stated power output of 135 W, is operated at 50% of full power. The "Biosonik," a generator manufactured by Bronwill Scientific Co., was the particular generator used. Under the above conditions no significant heating of the suspension occurs.

At this point in the procedure, a suspension of particles, suitably dispersed in a nonreactive fluid is available for further analysis. Ideally, the particles would be systematically separated into size groups and for each group the weight or particle number, the density range, and the abundance of each radioisotope would be determined. However, separations are hindered by loss of particles to the wall during the prolonged centrifugation periods required for systematic separation; the mass of dispersed fiber residue material interferes with direct weighing procedures and the submicron size of most of the particle population makes difficult the direct determination of specific activity and density. For these reasons, separate

particle samples are subjected to individual procedures designed to accomplish different parts of the analysis. These are described in the following paragraphs.

PARTICLE SIZE/NUMBER DETERMINATION BY MICROSCOPY

A uniform deposit of particles on a carbon coated electron microscope grid may be obtained by evaporating the fluid from a CCl_4 suspension in a beaker. A disk placed at the bottom of the beaker prior to ultrasonic dispersion of the filter has on its furface three circular areas milled to the depth of an electron microscope grid. Carbon covered grids are put in place when ultrasonic dispersion is complete. The carbon tetrachloride is evaporated under vacuum at room temperature to produce a uniform particle deposit over the grid areas. A number of grid fields are examined under optical and electron microscopes in order to find particle number per unit area for a series of particle size ranges. Data are used to obtain number versus size distribution curves. The characteristic spherical shape of active particles is used to distinguish these from fiber residue fragments which are irregular in shape.

SPECIFIC ACTIVITY OF PARTICLES BY DIRECT MEASUREMENT

The determination of the number of atoms of individual radionuclides per unit weight of particle is of considerable interest. The atoms of refractory radionuclide per microgram may be expected to be fairly constant with particle size, so that the refractory isotope can be used as a tracer for mass in a size distribution

determination. Experimentally, the problem is to separate a particle sample from inactive contaminants and then to determine weight and radioisotopic composition and abundance.

The radioactive spheres from a typical filter constitute perhaps 10% of the mass of the residue remaining after ashing, and to date, no effective method for separating the entire particle population from the fiber residue population has been found. However, it is possible to effect a bulk separation of the larger particles (those greater than about 1 to 2 μ in diameter) from the fiber residue. This may be accomplished by a series of 20-kc ultrasonic agitations carried out on 10-ml suspensions of the sample in CCl_4. Upon completion of the sonication period the larger particles are loosely clustered in a group at the bottom of the beaker in which the agitation occurred. The spherical particles of submicron dimension and the fiber residue particles are suspended in the fluid. The supernatant fluid may be gently decanted off to leave a large particle fraction in about 0.5 ml of CCl_4. Repeating this process three times produces a particle sample which is essentially fiber free and which may be weighed. Redistilled CCl_4 is used. The 0.5 ml of fluid and large particles remaining after the last decanting may be transferred by pipet and evaporated onto a previously tared aluminum weighing dish and the particle weight determined. A Cahn Electrobalance with recorder is used to obtain particle weights in the 5- to 20-μg range. After the weight is determined the entire weighing dish is mounted in the usual polyethylene cup for gamma spectroscopy.

An alternative method for separating the radioactive particles from filter residue depends on particle density. A density gradient from 3.25 to 2.00 g/cm^3 may be prepared in a 30-ml Beckman centrifuge tube using a continuously varying mixture of methyene iodide and carbon tetrachloride. A particle sample dispersed in CCl_4 is floated on top of the fluid column and the sample is centrifuged at 25,000 rpm overnight. The tube is sliced 1.5 cm from the base and the high density particles at the bottom recovered by filtering through a 0.22-μ pore size, 1-in. diam membrane filter. The filter is washed with CCl_4 and ignited to recover a weighable particle sample. However, the membrane filter ash weight must be corrected for and in general this method of separation does not produce as clean a particle sample as does the ultrasonic method.

SIZE SEPARATION AND PARTICLE DENSITY DETERMINATION

Particles produced by air detonations have densities which correspond to the oxides of the metals used in the delivery vehicle and are generally greater than 3 g/cm^3. There are no nonreactive organic fluids with high enough densities to use for density gradient separations at these density levels. However, particle density can be determined from Stokes' Law if the particle size and settling rate are known. During the size separation procedure, provision is made for obtaining particle microscopy samples at known settling times so that particle density determinations can be made.

Size separation of the small particles from airbursts is accomplished by sedimentation in a centrifugal field. The column used is like that described for gravitational sedimentation except that it is scaled down in size so that it can be centrifuged in a 50-ml swinging bucket system. The column is shown in Fig. 10.
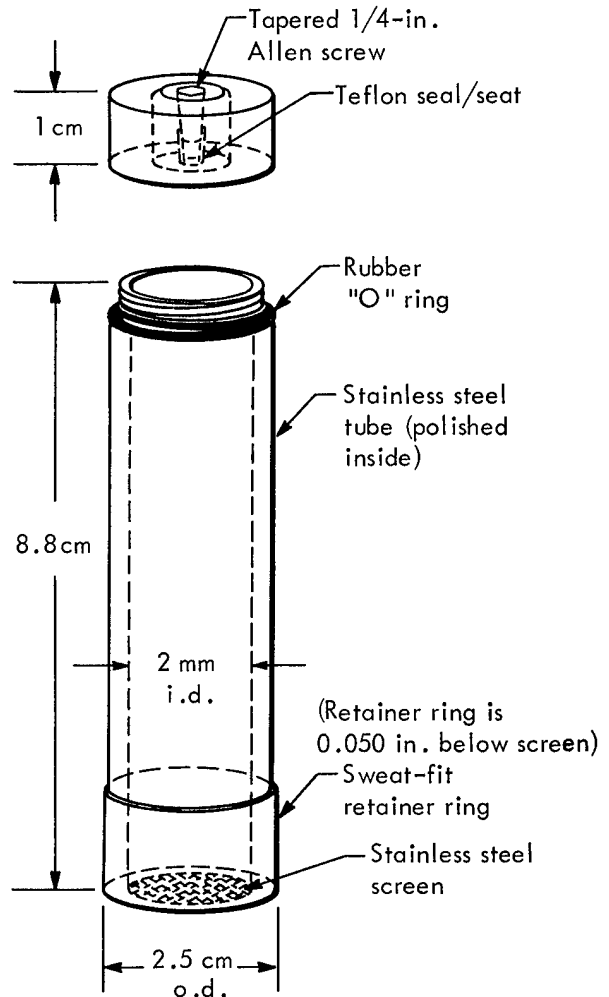

Fig. 10. Column for sedimentation in a centrifugal field.

Its operation is completely analogous to that of the gravitational column. It is sealed at the base with a rubber stopper on a Lab-Jack, and filled to within 1 cm of

the top with a 5 to 6% CCl_4 in 2-cSt silicone density gradient. The particle sample dispersed in 3 ml of 2-cSt silicone fluid is swirled briefly and poured down a stirring rod onto the top of the column. The column cap is placed in position and sealed. The rubber stopper base is removed and sample collection is begun.

The weight of particles collected in individual size fractions is of order of 1 μg or less and hence below the limits at which weight data can be obtained. Therefore, the size-separated fractions are collected primarily for gamma spectrometric analysis only and in order to minimize handling, fraction collections are made directly in the 1-in. diam polyethylene sample mounting cups. The larger size fractions are separated gravitationally, and smaller ones centrifugally, according to the sampling schedule described below. In all cases the sample is collected in 2 ml of 10% carbon tetrachloride in 2-cSt silicone fluid. To prepare the sample for counting, the collection fluid is evaporated in a drying oven at 100°C, the walls of the cup are washed with a minimum volume of CCl_4, and the fluid again evaporated. A lucite disk is placed in position to cover the sample and is glued to the wall of polyethylene cup.

The microscopy samples for density determination are taken in the interval between removal of one collection cup and positioning of the next. The microscopy sample is obtained in a small beaker containing about 1 ml of CCl_4. The surface of the CCl_4 is joined momentarily to the column base at the screen. Particles which have just reached the screen in the settling period are transferred to the CCl_4. By means of a transfer pipette, the $CCl_4/$ particle suspension is evaporated dropwise onto either a microscope slide or a carbon covered electron microscope grid for determination of particle diameter.

The flat bottom polyethylene cup used for fraction collection is supported during centrifugation by the flat surface of a solid aluminum hemisphere machined to fit exactly the bottom of the RC-2B centrifuge tube holder. (The tube holder delivered was slightly too small in diameter to accept the polyethylene cup freely, and the diameter was locally enlarged by about 0.002 in.) The base of the polyethylene cup supports the weight of the stainless steel screen bottom column during centrifugation. It is not affected at centrifugal speeds up to 5000 rpm; at higher speeds, centrifugation for prolonged periods causes the polyethylene to extrude away from the column base. For this reason centrifugal separation times are based on a maximum rate of 5000 rpm.

The relationship between particle size, particle density, and settling time in the stainless steel column for gravitational settling is:

$$M^2 t = \frac{\eta h}{3.27 \times 10^{-3} (d - \rho)} \qquad (23)$$

and for centrifugal settling it is:

$$M^2 t = \frac{\eta \, \ln R_2/R_1}{3.655 \times 10^{-2} (d - \rho)\left(\frac{rpm}{1000}\right)^2} \qquad (24)$$

In the above,

M = particle diameter in μ

t = settling time in min

d = particle density in g/cm^3

ρ = fluid density = 0.909 g/cm^3

η = fluid viscosity = 1.686 cP

R_2 = radial distance of screen from centrifuge axis = 12.8 cm

R_1 = radial distance of midpoint of sample from axis = 5.3 cm

h = $R_2 - R_1$ = 7.5 cm; $\ln R_2/R_1$ = 0.881

rpm = revolutions per minute.

Hence

$$M^2t(d - 0.909) = 3867 \text{ for gravitational settling} \qquad (25)$$

and

$$M^2t(d - 0.909) = 40.63/(rpm/1000)^2 \text{ for centrifugal.} \qquad (26)$$

A schedule of gravitational sedimentation times used together with the particle diameter data for an assumed 4.5 g/cm^3 density is given in Table II.

The sedimentation tube is then subjected to a series of centrifugation for the periods shown in Table III.

It should be noted that gravitational sampling times are cumulative while the centrifugal ones are incremental. In actual practice, the indicated times are corrected for start-up and shut-down of the centrifuge as well as for any significant gravitational settling that may have occurred between centrifugation periods.

$$t_{centrifugal} = \frac{0.0105}{(rpm/1000)^2} t_{gravitation}. \qquad (27)$$

The particle fractions collected by the above procedure are separated into size groups as indicated. However, the num-

Table II. Gravitational sedimentation in stainless steel column.

Time sample removed (min)	Diameter of particle at screen (μ)
15	8.4
30	6.0
60	4.2
120	3.0
240	2.1

Table III. Centrifugation sequence for stainless steel column.

Centrifugation time (min)	Centrifuge rpm	Diameter of particle at screen (μ)
2.5	1000	1.50
5.0	1000	1.06
10.0	1000	0.75
20.0	1000	0.53
5.0	2000	0.38
10.0	2000	0.26
20.0	2000	0.19
5.0	4000	0.13
10.0	4000	0.095
20.0	4000	0.065
25.6	5000	0.047
51.2	5000	0.032
102.4	5000	0.023
204.8	5000	0.016

ber distribution as a function of size may be biased by losses to the wall during the separation. Because of the small diameter of the tube, the fraction of particles lost to the wall during the prolonged sedimentation may be quite large. An alternate method which provides for the rapid separation of particle samples into two fractions—those less than and those greater than a given size—is based on a single centrifugal separation in a higher speed centrifuge. The Beckman Model L-2 Preparative Ultracentrifuge used operates a swinging bucket head at 20,000 rpm maximum speed.

The centrifuge takes a 1-in. diam, 3 in. high cellulose nitrate tube. A 5 to 6% CCl_4 in 2-cSt silicone density gradient column fills the tube to a height of 6 cm. The particle sample from a small section (usually one 7/8 in. disk) of IPC filter is ashed and dispersed in 2 ml of 2-cSt silicone as described above. The disperse suspension is poured onto the top of the column and the tube is immediately capped and transferred to the centrifuge. Centrifugation times calculated for particles of density 4.5 g/cm^3 traveling from the midpoint of the 1 cm thick sample layer to the point at which the tube will be sliced (1.55 cm from the base) are given by:

$$\frac{rpm^2}{1000} \times M^2 (\mu) \times t \text{ (min)} = 10.92$$

Thus, separation of particles greater than 1 μ in diameter from those less than 1 μ can be accomplished by 10.92-min centrifugation at 1000 rpm; separation of particles greater than 0.1 μ by 10.92-min centrifugation at 10,000 rpm; and separation of particles greater than 0.04 μ by

10.92 min at maximum speed, 25,000 rpm. Two component separation at 0.01-μ diam can be accomplished in 55 min at 25,000 rpm. Upon completion of centrifugation the tube is sliced at 1.55 cm from the base and the two solutions filtered onto 1-in. diam membrane filters or dropwise evaporated into polyethylene counting vials for gamma spectroscopy.

A series of two component separations of original sample can be used to provide data on the fraction of individual radioisotopes carried on particles of less than or greater than a series of particle sizes. These data can be used to determine radioisotopic distribution functions in the integral form.

SUMMARY—AERIAL SAMPLES FROM AIRBURSTS

(1) Filter paper is eliminated by ashing 4-7 hr in a muffle furnace at 400°C.

(2) Particles are removed from filter residue and dispersed by 20-kc ultrasonic agitation.

(3) Particle size distribution by number is determined by optical and electromicroscopic examination of particles uniformly deposited on electron microscope grid.

(4) Particle specific activity is determined by first isolating a weighable quantity of particles by means of a series of ultrasonic agitations and then determining the isotopic abundances by gamma spectroscopy.

(5) Size separation and particle density determination are accomplished by centrifugal sedimentation in a screen bottom tube to provide two series of particle

fractions—a series of samples is collected over time intervals of equal logarithmic increments for gamma spectroscopy; and in addition, the sedimentation fluid at the screen is sampled between collections to obtain samples for microscopic determination of particle size. These data can be used to determine particle density.

(6) Rapid, two component particle size separations are accomplished using an ultracentrifuge.

Gamma Spectrometry Procedures

All types of separated particle samples are mounted in 1-in. polyethylene cups. For each separation, a control sample consisting of an untreated portion of the original sample is similarly mounted. All gamma spectrometry is done with a lithium-drifted germanium diode detector. The detector is mounted so that the counting samples can be reproducibly positioned in close proximity to the diode. A cooled FET preamplifier is mounted in the vacuum. The pulse is divided as it comes out of the preamplifier and used to generate two spectra simultaneously—a low energy spectrum of 1024 channels at 0.25 keV per channel and a high energy spectrum of 2048 channels at 1 keV per channel. Both spectra are gain stabilized.

The division of the spectra into two regions provides for improved resolution in the low energy region (where photopeaks are numerous and closer together) while at the same time providing for coverage of the entire energy range of interest.

For a particular nuclear detonation, the spectra of the control samples are examined and compared with spectra of isotopic standards in order to identify each resolved photopeak. Based on these results, the one or two photopeaks which are best resolved for each isotope are selected for photopeak measurement. A simple computer program which sums the channels over the selected photopeak areas (and corrects for Compton continuum and background contributions) is used to process the spectra from all separated particle fractions from that event. That is, all fractions are assumed to have all isotopes present in the original material. Even through in some fractions the isotopic abundance of particular radioactive species might be quite low, this method produces meaningful results.

In most cases, it is sufficient to know only the relative distribution of the nuclides among the fractions and not their absolute amounts. In this case, resolved photopeak data require only correction for radioactive decay and the count/min in the photopeak can be used directly to obtain nuclide distribution with particle fraction. Absolute values can be obtained either by comparison with radioactive standards or by comparison with a control sample, a counterpart of which has previously been subjected to detailed radiochemical analysis.

The number of resolvable gamma emitting radioactive species decreases quite rapidly with sample age. However samples as old as twelve years still have four resolvable isotopes which cover the extreme range of fractionation behavior.

At twelve years, Cs^{137} which represents the extreme volatile behavior, Eu^{155} which represents the extreme refractory, and Sb^{125} and Co^{60}, both of which are intermediate in behavior, can all be measured by gamma spectroscopy. For more recent samples, after a delay of a few weeks, one can measure not only the above isotopes, but also up to 15 additional isotopes (depending on the event). For samples of this vintage, a single spectroscopic analysis provides all the data that can be obtained by gamma spectrometry. For very recent samples the photopeaks of short-lived species dominate the spectra at early times so that both an early spectrum and one taken after about two months contribute useful information. After gamma spectroscopy is completed, the sample may be recovered for microscopic or X-ray diffraction analysis.

In summary, the data taken on each particle fraction consists of sedimentation rate or density interval, and radioisotopic abundance, either in absolute or relative terms. Wherever possible both the weight and the chemical and physical nature of the separated particle fractions are determined. The object of obtaining these data is to arrive at a complete quantitative description of the overall particle population. The interpretation of the data for this purpose presents a different problem for each event analyzed and is discussed in the individual particle analysis reports for the events studied.

Acknowledgments

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